16th International Conference on Greenhouse Gas Control Technologies GHGT-16
23-27th October 2022, Lyon, France

CO2 mineralization in mafic rocks: from laboratory experiments to pilot site

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Abstract

Among the various CCS solutions considered to reduce CO2 emissions, mineral sequestration of CO2 offers a thermodynamically stable and reliable option over very long periods of time(1). However, the highest carbon mineralization potential is reached within specific environments, in particular mafic (basalts) and ultramafic rocks (peridotites, serpentinites, etc.). These lithologies are highly reactive and rich in magnesium, iron and calcium silicates (olivine, pyroxenes, plagioclases,...), and their efficiency in converting CO2 into carbonate minerals has been demonstrated by natural occurrences(2,3). Such rocks are present in many reservoirs: basaltic (e.g., Iceland, USA, India, Saudi Arabia), ophiolitic (Oman, New Caledonia, Liguria), but also in the form of mine tailings in the case of ex-situ sequestration (Canada, Australia, New Caledonia). Among these settings, 25 Gt of CO2 can theoretically be stored per year in mid-ocean ridge basaltic rocks(4).

Indeed, there are a number of scientific and technological hurdles to overcome in order to develop efficient and sustainable CO2 sequestration in these environments worldwide, which has been the focus of much of our research for over a decade. We are mainly focused on developing the knowledge necessary to make the transition from fundamental laboratory research to industrial applications within our various collaborations. Detailed studies are currently ongoing on i) abiotic and biotic reaction pathways of weathering and carbonation mechanisms in these rocks (thermodynamics and kinetics), ii) permeability variations, iii) measurements of various key parameters by complementary experimental methods, but also iv) monitoring and analysis of field data within the only pilot injection site in a mafic context in Iceland.

The study of the feasibility of forming carbonate minerals by injecting CO2 into basic rocks started in 2006-2008 with the first pilot site at Helligheidi in Iceland (CARBFIX and ANR-CO2FIX projects) (www.or.is/en/projects/carbfix5; www.anr.fr/Project-ANR-08-PCO2-0003). These first projects allowed a better quantification of the distribution of secondary mineral phases in fractures and their effects on permeability in many reservoirs. They also provided unique data on the stability and kinetics of carbonate minerals and the reactivity of deep ecosystems associated with basaltic rocks, their role in alteration and carbonate precipitation and the effects on CO2 injectivity6,7, which led to a second deeper injection study at industrial scale with the ongoing CARBFIX2 project (https://www.carbfix.com/carbfix2) and associated projects (Carnot ISIFoR, GECO: www.geco-h2020.eu).

In the MgO-CO2-H2O system, the precipitation of magnesite is very sluggish below 50 °C, which is mainly due to the highly hydrated character of the Mg2+ cation, and was confirmed by some recent studies of magnesite growth kinetics(8,9). As a result of its reluctance to dehydrate, magnesium has a strong tendency to form a variety of hydrated magnesium carbonates at ambient temperatures (hydromagnesite, nesquehonite and lansfordite, dypingite)10. In an attempt to bridge some of the existing gaps regarding the formation of magnesium-carbonates, we have been studying very thoroughly the solubility of carbonate minerals (6,11-16) and their precipitation rates, including near-equilibrium reactivity(17) and the effect of organic ligands on the solubility and precipitation rate of magnesite18.
In the CaO-CO₂-H₂O system, the precipitation of calcite and/or aragonite is relatively fast and efficient from ambient temperature to 170 °C\(^{19-22}\). Such carbon mineralization efficiency via Ca-carbonate formation was also confirmed by recent calculations on Ca isotopes from field injection fluids reacting with fresh basaltic rocks\(^{23}\). Yet, to expand the method applicability of fresh rocks towards commonly altered mafic reservoirs, the reactivity of a suite of secondary minerals (e.g., epidote, amphibole, chlorite, and serpentine) is currently under investigation\(^{24}\).

All these data are essential to determine not only the mechanisms that control mass transfers, but also to extrapolate the models resulting from the measurements carried out at different scales and to determine the injection and storage capacity of mafic rocks in the long term, as well as to dimension and optimize future industrial project.

Examples of carbon mineralization at different scales and geological context will be presented with their limitations and optimizations.

References

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Keywords: CO₂ mineralization; mafic rocks; pilot field site; experiments

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